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TITLE: Lightweight metal bipolar plates and methods for making the same

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Thin, light weight bipolar plates for use in electrochemical cells are rapidly, and inexpensively manufactured in mass production by die casting, stamping or other well known methods for fabricating magnesium or aluminum parts. The use of a light metal, such as magnesium or aluminum minimizes weight and simultaneously improves both electrical and thermal conductivity compared to conventional carbon parts. For service in electrochemical cells these components must be protected from corrosion. This is accomplished by plating the surface of the light weight metal parts with a layer of denser, but more noble metal. The protective metal layer is deposited in one of several ways. One of these is deposition from an aqueous solution by either electroless means, electrolytic means, or a combination of the two. Another is deposition by electrolytic means from a non-aqueous solution, such as a molten salt.

sheet material, smooth-surface channels, of substantially uniform depth, and having a clean, reverse image of the embossing die. Different flow field patterns and plate sizes will require different embossing pressures. The bulk of the sheet material (that is, the portions of the sheet material located apart from the channels) can also be compressed during the embossing operation and the embossing pressure can be selected to provide the appropriate channel depth in cross sectional profile, and also to impart the appropriate electrical conductivity and porosity to the bulk material."

# PATENT ABSTRACTS OF JAPAN

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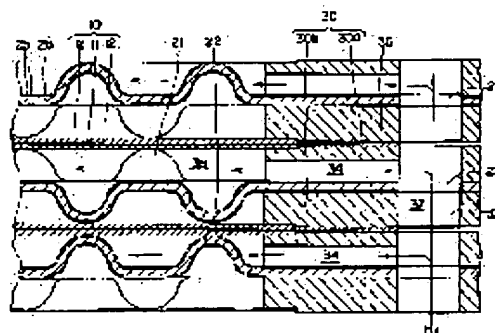
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## (54) SEPARATOR FOR FUEL CELL

### (57)Abstract:

**PURPOSE:** To provide a new structure of a separator for a fuel cell whose cost is reduced and which is excellent in productivity and whose safety is enhanced and which is excellent in supply efficiency of reaction gas.

**CONSTITUTION:** In a fuel cell stack formed by layering a plurality of cells 10 where electrodes 12 are arranged on both sides of solid electrolyte 11, it is used by being interposed between these cells. The obverse and the reverse of a metallic material excellent in workability are coated with a material excellent in electric conductivity, and a large number of projections 21 and 22 are arranged at proper intervals on these obverse and reverse. The projections are arranged so as to contact with cell surfaces of the fuel cell in the fuel cell stack. A space area 38 communicated and formed between the projections 21 between one side surface of a separator and the cells of the fuel cell becomes a fuel gas passing groove, and a space area communicated and formed between the projections 22 between the other side surface of the separator and the cells of the fuel cell becomes an oxidating agent gas passage groove.



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3. In the drawings, any words are not translated.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Industrial Application] this invention relates to the structure of the separator for fuel cells.

[0002]

[Description of the Prior Art] Various kinds, such as a solid-state polyelectrolyte type, a phosphoric-acid type, a melting carbonate type, and a solid-acid ghost type, are known according to the kind of electrolyte with which a fuel cell is used. Among these, it is a fuel cell using functioning as a solid-state polyelectrolyte type fuel cell carrying out the water of the macromolecule resin film which has a proton exchange group in a molecule to saturation as a proton conductivity electrolyte, and since it operates comparatively in the degree region of low temperature and the generating efficiency is also excellent, various kinds of uses are expected by making the object for electric vehicle loading into the start.

[0003] A solid-state polyelectrolyte type fuel cell stack has the structure which carried out the laminating of the fuel cell cell (single cell) which comes to join a gas diffusion electrode to both sides of a solid-state polyelectrolyte film by the means of a hotpress etc., and the gas separator made from a carbon metallurgy group (for example, refer to JP,6-119928,A).

[0004] The catalyst bed containing the catalyst active material allotted to the side to which a gas diffusion electrode touches an electrolyte film, While supporting this catalyst bed, reactant gas (fuel gas, oxidizer gas) is supplied and discharged. It consists of a gaseous diffusion layer of the porosity which furthermore also has a function as a charge collector. One gas diffusion electrode turns into a fuel electrode (anode pole) which receives supply of fuel gas (for example, gas which contains hydrogen gas or hydrogen in high concentration), and the gas diffusion electrode of another side turns into an oxidizer electrode (cathode pole) which receives supply of oxidizer gas (for example, air).

[0005] The example of composition of the fuel cell stack of two cells by such conventional technology is shown in drawing 5 - drawing 7 .

[0006] As for the single cell 10, it comes to join a gas diffusion electrode 12 to both sides of the electrolyte film 11 as mentioned above.

[0007] Separator supports and consists of states where it was contained inside the separator frame 4 with which the separator board 1 which consists of gas impermeability material (for example, precise carbon graphite) by which many concaves 2 and 3 were mutually formed in the front rear face in the rectangular direction, respectively consists of resin insulating materials, such as phenol resin. Opening formation of the manifold charge mouths 5a-5d for holding a gas manifold in the separator frame 4 is carried out.

[0008] In the fuel cell stack which comes to carry out the laminating of such separator and the single cell 10 6a is minded. the passage which fuel gas is introduced into the gas manifold (not shown) with which manifold charge mouth 5a is loaded, and is formed in the interior of seating-rim section 8a of the separator frame 4 -- a hole -- It is introduced into headroom field 7a between the separator board 1 and the separator frame 4, and the concave 2 by the side of the front face of the separator board 1 is flowed leftward in drawing 6 . and the passage formed in the interior of seating-rim section 8c which counters seating-rim section 8a -- it is discharged through a hole (not shown) by the fuel gas eccrisis manifold (not shown) with which manifold charge mouth 5b is loaded

[0009] The oxidizer gas which is the same as that of abbreviation also about oxidizer gas flowing, and was introduced into gas manifold section 5c 6b is minded. the passage formed in the interior of seating-rim section 8c of the separator frame 4 -- a hole -- A hole (not shown) is minded. the passage formed in the interior of 8d of seating-rim sections of the separator frame 4 after being introduced into lower part space field 7b between the separator board 1 and the separator frame 4 and flowing the concave 3 by the side of the rear face of the separator board 1 leftward in drawing 7 -- It is discharged by the oxidizer gas eccrisis manifold (not shown) with which 5d of manifold charge mouths is loaded.

[0010]

[Problem(s) to be Solved by the Invention] In the conventional separator as shown in drawing 5 - drawing 7 , although the concaves 2 and 3 of a large number used as a gas-passageway slot had to be formed in the front rear face, since the degree of hardness of the precise carbon graphite of separator material was very high, even if it used cutting tools, such as a diamond tool, cutting was not easy, and there was a problem that mass production was difficult.

[0011] Moreover, on especially the oxidizer pole, although the generation water by the cell reaction needed to be discharged efficiently, the gas-passageway slots of the conventional separator were two or more parallel slots, and generation water tended to pile up and they were missing at eccrisis efficiency. When the eccrisis efficiency of generation water was bad, it became the

cause by which an electrode exfoliated from an electrolyte film, and reactant gas reacted with the catalyst on an electrode, and the danger of producing the trouble of igniting in an electrode edge was latent.

[0012]

[Means for Solving the Problem] then, this invention canceled the trouble of the above-mentioned conventional technology, its productivity was good at the low cost, and its safety was high and, moreover, excellent in the supply efficiency of reactant gas -- it aims at offering the structure of the new separator for fuel cells

[0013] Moreover, this invention is combined and aims at miniaturizing and having separator and attaining small lightweight-ization of a fuel cell stack.

[0014] In order to attain these purposes, this invention is inserted between the aforementioned fuel cell cells in the fuel cell stack to which it comes to carry out two or more laminatings of the fuel cell cell which allotted the electrode to the both sides of a solid electrolyte, and is used. While while adjoins one side and having a fuel gas passage slot for supplying fuel gas to a fuel cell cell It is the separator for fuel cells which equipped the fuel cell cell of another side which adjoins the side of another side with the oxidizer gas-passageway slot for supplying oxidizer gas. The front rear face of the metallic material excellent in processability is coated with a material excellent in electrical conductivity. And much salients allot a suitable interval to the front rear face, and are prepared in it, respectively. It is prepared and the aforementioned salient becomes so that the aforementioned fuel cell cell side may be touched in the aforementioned fuel cell stack, and it is characterized by carrying out free passage formation of the aforementioned fuel gas passage slot and the aforementioned oxidizer gas-passageway slot between the aforementioned salients between the aforementioned separator and the aforementioned fuel cell cell, respectively.

[0015]

[Function] The fuel gas supplied to separator from the gas manifold with which the gas manifold charge mouth for fuel gas supply by which opening formation is carried out is loaded passes along the fuel gas passage slot by which free passage formation is carried out between the salients of a separator side between separator and a fuel cell cell in one separator side, and is discharged in the gas manifold with which the gas manifold charge mouth for opening formation fuel gas eccrisis is loaded like the opposite side of separator. Oxidizer gas passes along the oxidizer gas-passageway slot by which free passage formation is similarly carried out between the salients of a separator side from an oxidizer supply gas manifold at the another side side of separator, and is discharged in an oxidizer eccrisis gas manifold.

[0016]

[Example] With reference to drawing 1 or drawing 4 , the composition of the separator for fuel cells by one example of this invention is explained below.

[0017] the separator frame of a couple with which this separator serves as the separator board 20 from resin insulating materials, such as phenol resin, -- it consists of separator frames 30 which come to join Members 30a and 30b the separator board 20 -- a separator frame -- pinching fixation is carried out among Members 30a and 30b

[0018] The metallic material with easy embossing or dimple processing for the separator board 20 with reference to drawing 1 , SUS, cold rolling material, aluminum, etc. are more specifically made into a base material. at the front rear face Gas impermeability material with good electrical conductivity, For example, sinking in, thermal spraying, electrodeposition, sputtering, etc. coat precise carbon graphite by technique suitably, embossing or dimple processing is given to this, and much salients 21 and 22 are formed in the front rear face at intervals of several mm. When a fuel cell stack is constituted, the height to the summit of salients 21 and 22 is set up so that the summit of a salient may be close to the fuel cell single cell 10 ( drawing 3 ).

[0019] In addition, after coating a substrate table rear face with gas impermeability material, embossing or dimple processing may be given, and after giving embossing or dimple processing to a substrate table rear face and forming salients 21 and 22 in it on the contrary, you may coat gas impermeability material.

[0020] Penetration formation of the manifold charge mouth 23 for loading with a gas manifold, respectively is carried out on the 4 rounds of outsides of the field in which salients 21 and 22 were formed. Moreover, penetration formation of the breakthrough 24 for making the bolt or tie rod for stack fixation penetrate squarely is carried out.

[0021] the separator frame 30 -- the separator frame of the same composition -- it is formed by joining Members 30a and 30b A sealing compound is beforehand applied to a part for each joint.

[0022] a separator frame -- a member -- the composition of 30a (30b) is shown in drawing 2 a separator frame -- a member -- penetration formation of the opening 31 is carried out in the center of 30a Penetration formation of the manifold charge mouth 32 for loading 4 rounds of outsides of the central opening 31 with a gas manifold, respectively is carried out, and the breakthrough 33 for making the bolt or tie rod for stack fixation penetrate squarely is formed. These are prepared so that it may align, respectively with the manifold charge mouth 23 in the separator board 20, and a breakthrough 24.

[0023] a separator frame -- a member -- the inferior-surface-of-tongue side of 30a -- setting -- many gas passageways -- a hole 34 counters and is prepared these gas passageways -- in the ends, opening of the hole 34 is carried out to the manifold charge mouths 32 and 32 and the central opening 31, respectively

[0024] the separator frame constituted as mentioned above -- oppose the upper surface which changes the sense 90 degrees and is shown in drawing 2 as a rectangular state in Members 30a and 30b, respectively -- alike -- carrying out -- these separator frame -- the separator of this example is constituted by making it join mutually on both sides of the separator board 20 among Members 30a and 30b

[0025] When two single cells 10 and 10 are pinched between separator, respectively and the fuel cell stack of two cells is constituted using such separator, on the other hand, Mukai's cross section is shown in drawing 3 . Each fuel cell single cell 10 comes to join gas diffusion electrodes 12 and 12 to both sides of the solid-state polyelectrolyte film 11 by the means of a

hotpress etc. the edge of the electrolyte film [ in / the fuel cell single cell 10 / on between each separator, and ] 11 -- a separator frame -- it is pinched among Members 30a and 30b, and adhesion fixation is carried out by the sealing compound 36 by thermosetting resin, such as an epoxy resin

[0026] drawing 3 -- setting -- a sign 37 -- the charge field for a fuel gas introduction manifold (not shown) -- being shown -- the separator board 20 and a separator frame -- one each of each of the manifold charge mouths 23 and 32 formed in Members 30a and 30b is making this manifold charge field 37 in line The fuel gas introduced into the fuel gas introduction manifold A passage the separator frame of the manifold charge mouth 37 to a top -- a member -- the gas passageway prepared in the undersurface side of 30a -- a hole 34 Furthermore, it flows in the direction of an arrow through the space field 38 currently formed between the upper surface of the separator board 20, and the undersurface of the single cell 10 succeeding between the salients 21 of a large number formed in the separator board upper surface. It is discharged by the fuel gas discharge manifold (not shown) with which the manifold charge mouth of an opposite side is loaded.

[0027] The cross section of the direction which intersects perpendicularly is indicated to be drawing 3 of the above-mentioned fuel cell stack to drawing 4 . In drawing 4 , a sign 39 shows the charge field for an oxidizer gas introduction manifold (not shown). The oxidizer gas introduced into the oxidizer gas introduction manifold A passage the separator frame of the manifold charge mouth 39 to the bottom -- a member -- the gas passageway prepared in the upper surface side of 30b -- a hole 35 Furthermore, it flows in the direction of an arrow through the space field 40 currently formed between the separator board 20 and the single cell 10 succeeding between much salients 22, and is discharged by the oxidizer gas discharge manifold (not shown) with which the manifold charge mouth of an opposite side is loaded.

[0028]

[Effect of the Invention] Since according to this invention embossing or dimple processing gave embossing or dimple processing to this front rear face, formed much salients in it, using as separator what coated the front rear face of an easy metallic material with a gas impermeability material excellent in electrical conductivity and considered as the reactant gas passage slot, processing of a reactant gas passage slot is easy, and it is possible to mass-produce separator in a low cost.

[0029] Moreover, in the reactant gas passage slot which consists of the conventional parallel slot, reactant gas supply efficiency can improve according to the turbulent flow effect which was not acquired, and stay of the generation water in an oxidizer pole can also be canceled.

[0030] Furthermore, according to this invention for the thickness of 5.3mm of the separator by conventional precise carbon graphite, it is possible to about 0.5mm to make it thin. For this reason, large small lightweight-ization is realized in the fuel cell stack to which it comes to carry out the laminating of many fuel cell cells and separator.

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[Translation done.]

## Bipolar metallic plate for high-temperature fuel cell stack

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Applicant(s): SIEMENS AG (DE)  
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EC Classification: [H01M8/02C2A](#)  
Equivalents:  
Cited Documents: [DE19541187](#); [WO9723006](#); [DE4410711](#); [JP62262376](#); [JP2132764](#); [JP6052868](#); [JP6260178](#)

### Abstract

A high temperature fuel cell (4, 5) has chromium-containing interconnector plates (6) which bear, on the cathode-facing side, a coating (28) comprising a surface layer (30) of nickel and an overlying layer (32) of aluminium. Preferably, the surface layer (30) is a 10-100  $\mu$  thick layer containing  $\geq$  90 wt.% Ni and the overlying layer (32) is a 10-20  $\mu$  thick layer containing  $\geq$  90 wt.% Al. The coating is preferably provided on the side walls (24) and bases (26) of parallel channels in the plate surface (16). Also claimed is a high temperature fuel cell stack (2) with several of the above fuel cells (4, 5).

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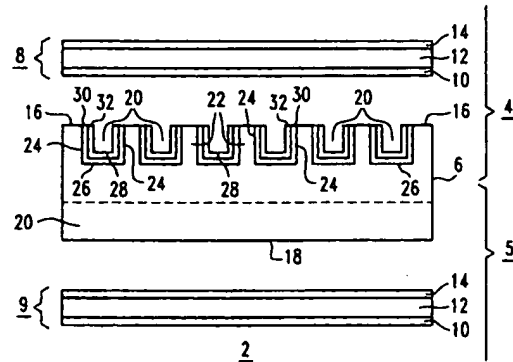
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(54) **Metallische bipolare Platte für Hochtemperatur-Brennstoffzellenstapel**

(57) Bei der vorliegenden Hochtemperatur-Brennstoffzelle (4,5) mit wenigstens einer chromhaltigen Verbundleiterplatte (6), die auf der der Kathode (6) zugewandten Oberfläche (16) wenigstens teilweise eine Schicht (28) aufweist, umfaßt die Schicht (28) wenigstens eine erste und eine zweite Lage (30,32). Die erste, auf der Oberfläche (16) der Verbundleiterplatte (6) angeordnete Lage (30), enthält Nickel (Ni) und die darüber angeordnete zweite Lage (32) Aluminium (Al). Durch diese Maßnahme wird das Verdampfen von chromhaltigen Verbindungen aus der Verbundleiterplatte (6) weitgehend vermieden.





## Beschreibung

Die Erfindung bezieht sich auf eine Hochtemperatur-Brennstoffzelle und auf einen Hochtemperatur-Brennstoffzellenstapel.

Es ist bekannt, daß bei der Elektrolyse von Wasser die Wassermoleküle durch elektrischen Strom in Wasserstoff ( $H_2$ ) und Sauerstoff ( $O_2$ ) zerlegt werden. In einer Brennstoffzelle läuft dieser Vorgang in umgekehrter Richtung ab. Durch die elektrochemische Verbindung von Wasserstoff ( $H_2$ ) und Sauerstoff ( $O_2$ ) zu Wasser entsteht mit hohem Wirkungsgrad elektrischer Strom. Dies geschieht, wenn als Brenngas reiner Wasserstoff ( $H_2$ ) eingesetzt wird, ohne Emission von Schadstoffen und Kohlendioxid ( $CO_2$ ). Auch mit einem technischen Brenngas, beispielsweise Erdgas oder Kohlegas, und mit Luft (die zusätzlich mit Sauerstoff ( $O_2$ ) angereichert sein kann) anstelle von reinem Sauerstoff ( $O_2$ ) erzeugt eine Brennstoffzelle deutlich weniger Schadstoffe und weniger Kohlendioxid ( $CO_2$ ) als andere Energieerzeuger, die mit fossilen Energieträgern arbeiten. Die technische Umsetzung des Prinzips der Brennstoffzelle hat zu unterschiedlichen Lösungen, und zwar mit verschiedenartigen Elektrolyten und mit Betriebstemperaturen zwischen 80 °C und 1000 °C, geführt.

In Abhängigkeit von ihrer Betriebstemperatur werden die Brennstoffzellen in Nieder-, Mittel- und Hochtemperatur-Brennstoffzellen eingeteilt, die sich wiederum durch verschiedene technische Ausführungsformen unterscheiden.

Bei dem aus einer Vielzahl von Hochtemperatur-Brennstoffzellen sich zusammensetzenden Hochtemperatur-Brennstoffzellenstapel (in der Fachliteratur wird ein Brennstoffzellenstapel auch "Stack" genannt) liegen unter einer oberen Verbundleiterplatte, welche den Hochtemperatur-Brennstoffzellenstapel abdeckt, der Reihenfolge nach wenigstens eine Schutzschicht, eine Kontaktschicht, eine Elektrolyt-Elektroden-Einheit, eine weitere Kontaktschicht, eine weitere Verbundleiterplatte, usw.

Die Elektrolyt-Elektroden-Einheit umfaßt zwei Elektroden und einen zwischen den beiden Elektroden angeordneten, als Membran ausgeführten Festelektrolyten. Dabei bildet jeweils eine zwischen benachbarten Verbundleiterplatten liegende Elektrolyt-Elektroden-Einheit mit den beidseitig an der Elektrolyt-Elektroden-Einheit unmittelbar anliegenden Kontaktschichten eine Hochtemperatur-Brennstoffzelle, zu der auch noch die an den Kontaktschichten anliegenden Seiten jeder der beiden Verbundleiterplatten gehören. Dieser Typ und weitere Brennstoffzellen-Typen sind beispielsweise aus dem "Fuel Cell Handbook" von A. J. Appleby und F. R. Foulkes, 1989, Seiten 440 bis 454, bekannt.

Die metallischen Verbundleiterplatten in der Hochtemperatur-Brennstoffzelle (und damit auch die im Hochtemperatur-Brennstoffzellenstapel) bestehen vorzugsweise aus einer Eisenbasislegierung oder einer

Chrombasislegierung (bei einer Basislegierung besteht der Werkstoff zu wenigstens 50 Gew.-% aus dem in der Basislegierung genannten Element). Aufgrund der hohen Betriebstemperatur (von beispielsweise über 600 °C) beim Betrieb der Hochtemperatur-Brennstoffzelle ist die Zusammensetzung der jeweiligen Basislegierung dergestalt zusammenzusetzen, daß sich während des Betriebes eine Korrosionsschutzschicht auf der der Kathode zugewandten Oberfläche der Verbundleiterplatte bildet. Es kommt beispielsweise eine Korrosionsschutzschicht aus Chromoxid ( $Cr_2O_3$ ) in Betracht, da diese neben der Schutzwirkung auch noch eine ausreichende elektrische Leitfähigkeit aufweist.

Wenn der Werkstoff der Verbundleiterplatte größere Mengen Chrom enthält (z.B. aus einer Chrombasislegierung besteht), bildet sich die Korrosionsschutzschicht der Verbundleiterplatte bereits schon bei sehr niedrigen Sauerstoffpartialdrücken. Dies hat jedoch den unerwünschten Nebeneffekt, daß flüchtige Chromverbindungen aus der Verbundleiterplatte abdampfen können. Dadurch wird die Funktionsfähigkeit der Hochtemperatur-Brennstoffzelle beeinträchtigt und damit auch die Leistungsfähigkeit des gesamten Hochtemperatur-Brennstoffzellenstapels. Die Einschränkung der Leistungsfähigkeit ist hier durch Alterungseffekte in den Elektroden der Elektrolyt-Elektroden-Einheit bedingt.

Aus der deutschen Patentschrift 44 10 711 ist eine Verbundleiterplatte aus einer Chrombasislegierung bekannt, die auf ihrer der Kathode (d.h. auf der sauerstoffführenden Seite) zugewandten Seite eine Schutzschicht aus Aluminiumoxid ( $Al_2O_3$ ) aufweist. Diese Schutzschicht wird unmittelbar auf die chromhaltige Verbundleiterplatte aufgetragen, mit dem Ziel, daß flüchtige Chromverbindungen nun nicht mehr aus der Verbundleiterplatte abdampfen können.

Versuche haben jedoch gezeigt, daß Anteile der aufgedampften Schutzschicht aus Aluminiumoxid ( $Al_2O_3$ ) nach dem Auftragen in den Werkstoff der Verbundleiterplatte eindiffundieren. Aufgrund dieses Materialschwunds in der Schutzschicht verliert diese ihre schützende Wirkung gegen das Verdampfen von chromhaltigen Verbindungen aus der Verbundleiterplatte. In der Schutzschicht sind Rißbildungen zu beobachten (die Porosität der Schutzschicht nimmt im starken Maße zu), welche ein Abdampfen der chromhaltigen Verbindungen aus der Verbundleiterplatte ermöglichen. Der gewünschte Effekt, daß die Schutzschicht aus Aluminiumoxid ( $Al_2O_3$ ) über eine längere Betriebsdauer das Verdampfen aus der chromhaltigen Verbundleiterplatte unterbindet, ist somit nicht gewährleistet.

Der Erfindung liegt nun die Aufgabe zugrunde, eine Hochtemperatur-Brennstoffzelle und einen Hochtemperatur-Brennstoffzellenstapel dergestalt anzugeben, daß das Verdampfen von chromhaltigen Verbindungen aus einer chromhaltigen Verbundleiterplatte weitgehend vermieden wird.

Bei einer Hochtemperatur-Brennstoffzelle mit wenigstens einer chromhaltigen Verbundleiterplatte, weist die Verbundleiterplatte auf der der Kathode zugewandten Oberfläche wenigstens teilweise eine Schicht auf, die gemäß der Erfindung wenigstens eine erste und eine zweite Lage umfaßt, wobei die erste, auf der Oberfläche der Verbundleiterplatte angeordnete Lage, Nickel (Ni) und die darüber angeordnete zweite Lage Aluminium (Al) enthält.

Durch diese zweilagige Schicht ist gewährleistet, daß ein Verdampfen von chromhaltigen Verbindungen aus der chromhaltigen Verbundleiterplatte weitgehend vermieden wird. Dadurch wird die Funktionsfähigkeit der Hochtemperatur-Brennstoffzelle und zugleich die Leistungsfähigkeit derselben verbessert. Die zuerst auf die Oberfläche der Verbundleiterplatte aufgetragene nickelhaltige Schicht diffundiert teilweise in den chromhaltigen Werkstoff der Verbundleiterplatte ein. Es bildet sich eine stabile (verzahnende) Verbindung zwischen der ersten Lage und der Verbundleiterplatte. Auf diese erste Lage wird darüber die zweite aluminiumhaltige Lage aufgetragen. Da diese entgegen dem Stand der Technik nicht unmittelbar auf die Verbundleiterplatte aufgetragen wird, dringt diese aluminiumhaltige Lage nicht in den Werkstoff der Verbundleiterplatte ein. Die nickelhaltige erste Lage hat praktisch eine sperrende Wirkung gegenüber der aluminiumhaltigen zweiten Lage bezüglich des teilweisen Eindringens (d.h. des Eindiffundierens) in die Verbundleiterplatte. Das Aluminium der zweiten Lage dringt nur noch in die nickelhaltige erste Lage (die sogenannten Sperrlage) ein und wird dort abgebunden. Im Grenzbereich zwischen der nickelhaltigen ersten Lage und der aluminiumhaltigen zweiten Lage (d.h. bis zu einer bestimmten Tiefe in der ersten und der zweiten Lage) bilden sich Mischkristalle aus Aluminium (Al) und Nickel (Ni). Diese Mischkristalle (z.B.  $\text{Ni}_3\text{Al}$ ) können zwischen den Schichten eine intermetallische Phase bilden oder als Sekundärausscheidungen in den Schichten eingelagert werden.

Während des Betriebes der Hochtemperatur-Brennstoffzelle bildet sich auf der der Kathode zugewandten Seite der aluminiumhaltigen Lage eine Aluminiumoxidschicht ( $\text{Al}_2\text{O}_3$ ). Die zweilagige Schicht verhindert also das Verdampfen von chromhaltigen flüchtigen Verbindungen aus der Verbundleiterplatte.

Vorzugsweise enthält die erste Lage wenigstens 90 Gew.-% Nickel. Dadurch ist gewährleistet, daß die nickelhaltige erste Lage eine gute Haftung auf der Oberfläche der Verbundleiterplatte besitzt und zugleich das Aluminium (Al) aus der aluminiumhaltigen zweiten Lage nicht in die Verbundleiterplatte diffundiert. Das Aluminium wird in der nickelhaltigen ersten Lage gestoppt (mit anderen Worten gesperrt).

Insbesondere kann die erste Lage eine Dicke zwischen 10 und 100  $\mu\text{m}$  besitzen. Dieser Bereich für die Dicke hat sich experimentell zum Verhindern (zum Sperren) der Diffusion von Aluminium aus der zweiten Lage in die Verbundleiterplatte bewährt.

In einer weiteren Ausgestaltung enthält die zweite Lage wenigstens 90 Gew.-% Aluminium und besitzt eine Dicke zwischen 10 und 20  $\mu\text{m}$ . Diese Wertebereiche für die Konzentration und die Dicke der zweiten Lage reichen aus, das Verdampfen von chromhaltigen Verbindungen aus der Verbundleiterplatte zu unterbinden.

Bei der Hochtemperatur-Brennstoffzelle verlaufen in der Regel wenigstens zwei Kanäle nebeneinander in der Oberfläche der Verbundleiterplatten, wobei jeweils zwei Kanäle durch einen Steg voneinander getrennt sind; dabei sind vorzugsweise die Seitenwände und die Böden der Kanäle mit der Schicht versehen. Damit die Schicht elektrisch isolierend wirkt, werden die Oberflächen der Stege nicht mit der Schicht versehen. Durch die Oberflächen der Stege fließt senkrecht zu diesen der elektrische Strom.

Gemäß der Erfindung weist ein Hochtemperatur-Brennstoffzellenstapel eine Anzahl solcher Hochtemperatur-Brennstoffzellen auf.

Weitere vorteilhafte Ausgestaltungen sind in den Unteransprüchen wiedergegeben. Zum besseren Verständnis der Erfindung und ihrer Weiterbildungen werden zwei Ausführungsbeispiele anhand einer Figur erläutert. Die Figur zeigt einen Ausschnitt aus einem Hochtemperatur-Brennstoffzellenstapel mit zwei Hochtemperatur-Brennstoffzellen in schematischer Darstellung.

In der Figur erkennt man einen Ausschnitt aus einem Hochtemperatur-Brennstoffzellenstapel 2. Der Ausschnitt umfaßt zwei nicht vollständig dargestellte Hochtemperatur-Brennstoffzellen 4,5 (in der Regel enthält ein Hochtemperatur-Brennstoffzellenstapel wenigstens fünfzig Hochtemperatur-Brennstoffzellen).

Die Hochtemperatur-Brennstoffzellen 4,5 umfassen jeweils in der angegebenen Reihenfolge eine Verbundleiterplatte 6 (innerhalb eines Hochtemperatur-Brennstoffzellenstapels, d.h. nicht an seinen Enden, wird die Verbundleiterplatte 6 auch als bipolare Platte bezeichnet), sowie eine Elektrolyt-Elektroden-Einheit 8,9 und eine weitere nicht dargestellte Verbundleiterplatte. Zwischen den Verbundleiterplatten 6 und den Elektrolyt-Elektroden-Einheiten 8,9 werden in der Regel außerdem noch zusätzliche Kontaktschichten angeordnet.

Die Elektrolyt-Elektroden-Einheiten 8,9 umfassen in der angegebenen Reihenfolge jeweils eine Kathode 10, einen Festelektrolyten 12 und eine Anode 14.

Die Verbundleiterplatte 6 besteht aus einer Chrombasislegierung. In die Oberfläche 16, die der Kathode 10 der Elektrolyt-Elektroden-Einheit 8 zugewandt ist, und in die Oberfläche 18, die der Anode 14 der Elektrolyt-Elektroden-Einheit 9 zugewandt ist, sind parallel zueinander verlaufende Kanäle 20 eingearbeitet. Jeweils zwei parallel zueinander verlaufende Kanäle 20 sind durch einen Steg 22 voneinander getrennt. In den Kanälen 20 werden Betriebsmittel zum Betreiben des Hochtemperatur-Brennstoffzellenstapels 2 geführt. In den Kanälen 20, die der Kathode 10 der Elektrolyt-Elek-

troden-Einheit 8 zugewandt sind, wird beispielsweise Sauerstoff (O<sub>2</sub>) geführt, wohingegen in den Kanälen 20, die der Anode 14 der Elektrolyt-Elektroden-Einheit 9 zugewandt sind, Wasserstoff (H<sub>2</sub>) geführt wird.

Die Kanäle 20 in der Oberfläche 16 sind auf ihren Seitenflächen 24 und auf ihren Böden 26 mit einer zweilagigen Schicht 28 versehen. Die Schicht 28 umfaßt eine erste Lage 30 und eine zweite Lage 32. Die erste Lage 30 enthält Nickel (Ni) und ist unmittelbar auf der Oberfläche 16 (genauer: auf den Seitenflächen 24 und den Böden 26 der Kanäle 20) angeordnet. Die zweite Lage 32 ist über der ersten Lage 30 angeordnet und enthält Aluminium (Al).

In einem weiteren nicht dargestellten Ausführungsbeispiel sind auch die Stege 22 auf ihren Oberflächen mit der Schicht 28 versehen. Eine Maske (zum Abdecken der Oberflächen der Stege 22) wird dann nicht mehr benötigt. Allerdings muß die nichtleitende Schicht 28 in einem weiteren Schritt von den Oberflächen der Stege 22 entfernt werden.

Mit der Schicht 28 wird nun erreicht, daß während des Betriebes des Hochtemperatur-Brennstoffzellenstapels 2 (und damit der Hochtemperatur-Brennstoffzellen 4,5) das Verdampfen von chromhaltigen Verbindungen aus der Verbundleiterplatte 6 verhindert wird.

Die erste Lage 30 enthält wenigstens 90 Gew.-% Nickel (Ni) und besitzt vorzugsweise eine Dicke zwischen 10 und 100 µm. Die zweite Lage 32 enthält wenigstens 90 Gew.-% Aluminium (Al) und weist insbesondere eine Dicke zwischen 10 und 20 µm auf. Der Werkstoff der nickelhaltigen ersten Lage 30 dringt teilweise in die Oberfläche 16 (d.h. hier in die Seitenwände 24 und die Böden 26) der Kanäle 20 ein und gewährleistet eine stabile (verzahnende) Verbindung zwischen der Verbundleiterplatte 6 und der aluminiumhaltigen zweiten Lage 32. Die erste Lage 30 verhindert das Eindringen (das Hineindiffundieren) des Aluminiums (Al) der aluminiumhaltigen zweiten Lage 32 in die Verbundleiterplatte 6 (die erste Lage 30 hat also die Funktion einer Sperrschicht für das Aluminium (Al)).

Das Aluminium (Al) dringt nun nur noch in die erste Lage 30 ein und bildet mit dem Nickel (Ni) der ersten Lage 30 einen Mischkristall (eine intermetallische Phase zwischen den Metallen Nickel (Ni) und Aluminium (Al)).

Die zweilagige Schicht 28, die die erste Lage 30 und die zweite Lage 32 umfaßt, weist eine glatte feinporeige Oberfläche aus. Während des Betriebes bildet sich an der der Kathode 10 zugewandten Oberfläche der zweiten Lage 32 eine Aluminiumoxid (Al<sub>2</sub>O<sub>3</sub>)-Schicht. Das Verdampfen von chromhaltigen Verbindungen aus der chromhaltigen Verbundleiterplatte 6 wird während des Betriebes des Hochtemperatur-Brennstoffzellenstapels 2 weitgehend vermieden. Dadurch kommt es nicht zu Alterungserscheinungen in den Kathoden 10 der Elektrolyt-Elektroden-Einheiten 8,9.

Eine Funktionseinschränkung des Hochtempera-

tur-Brennstoffzellenstapels 2 aufgrund der Verdampfung von chromhaltigen Verbindungen aus den chromhaltigen Verbundleiterplatten 6 ist somit nahezu ausgeschlossen. Die Leistungsfähigkeit des gesamten Hochtemperatur-Brennstoffzellenstapels 2 wird somit verbessert.

Auf der der Anode 14 der Elektrolyt-Elektroden-Einheit 9 zugewandten Oberfläche 18 der Verbundleiterplatte 6 wird kein Sauerstoff (O<sub>2</sub>) als Betriebsmittel geführt, so daß es nicht zu unerwünschten Korrosionserscheinungen und zum Verdampfen von chromhaltigen Verbindungen aus der Verbundleiterplatte 6 kommen kann. Die Schicht 28 wird somit auf der Oberfläche 18 der Verbundleiterplatte 6 nicht benötigt.

## Patentansprüche

1. Hochtemperatur-Brennstoffzelle (4,5) mit wenigstens einer chromhaltigen Verbundleiterplatte (6), wobei die Verbundleiterplatte (6) auf der der Kathode (6) zugewandten Oberfläche (16) wenigstens teilweise eine Schicht (28) aufweist, **dadurch gekennzeichnet**, daß die Schicht (28) wenigstens eine erste und eine zweite Lage (30,32) umfaßt, wobei die erste, auf der Oberfläche (16) der Verbundleiterplatte (6) angeordnete Lage (30), Nickel (Ni) und die darüber angeordnete zweite Lage (32) Aluminium (Al) enthält.
2. Hochtemperatur-Brennstoffzelle (4,5) nach Anspruch 1, **dadurch gekennzeichnet**, daß die erste Lage (30) wenigstens 90 Gew.-% Nickel (Ni) enthält.
3. Hochtemperatur-Brennstoffzelle (4,5) nach Anspruch 1 oder 2, **dadurch gekennzeichnet**, daß die erste Lage (30) eine Dicke zwischen 10 und 100 µm besitzt.
4. Hochtemperatur-Brennstoffzelle (4,5) nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet**, daß die zweite Lage (32) wenigstens 90 Gew.-% Aluminium (Al) enthält.
5. Hochtemperatur-Brennstoffzelle (4,5) nach einem der vorhergehenden Ansprüche, **dadurch gekennzeichnet**, daß die zweite Lage (32) eine Dicke zwischen 10 und 20 µm besitzt.
6. Hochtemperatur-Brennstoffzelle (4,5) nach einem der vorhergehenden Ansprüche mit wenigstens zwei in der Oberfläche (16) parallel zueinander verlaufenden Kanälen (20), wobei jeweils zwei Kanäle (20) durch einen Steg (22) voneinander getrennt sind, **dadurch gekennzeichnet**, daß die Seitenwände (24) und die Böden (26) der Kanäle (20) mit der Schicht (28) versehen sind.

7. Hochtemperatur-Brennstoffzellenstapel (2) mit einer Anzahl von Hochtemperatur-Brennstoffzellen (4,5) nach einem der Ansprüche 1 bis 6.

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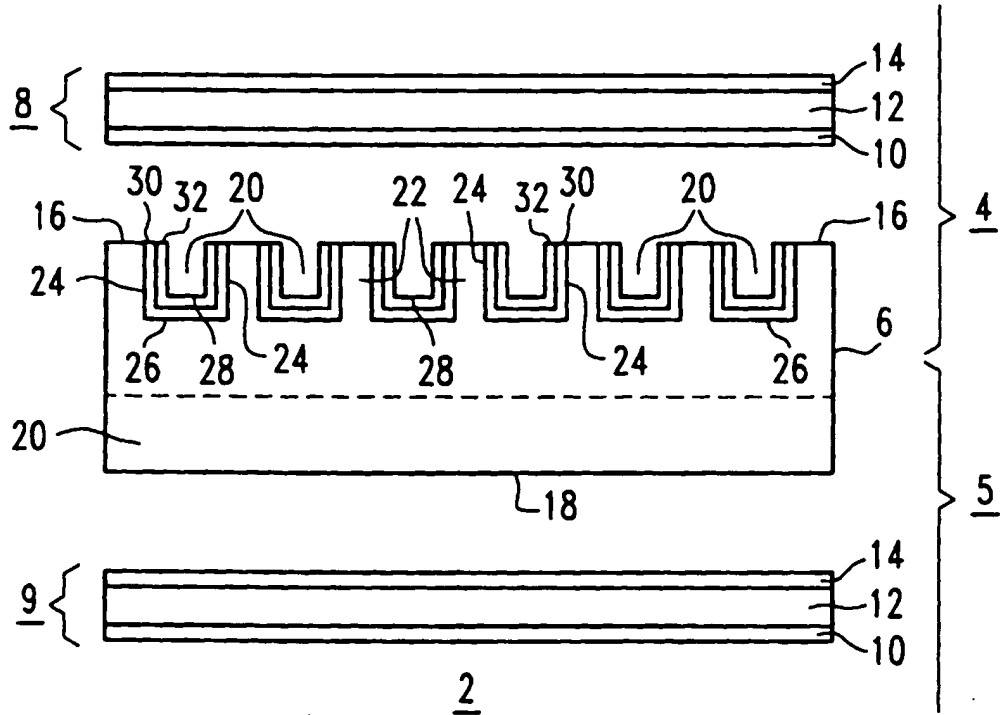
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Europäisches  
Patentamt

# EUROPÄISCHER RECHERCHENBERICHT

Nummer der Anmeldung  
EP 97 11 1207

EINSCHLÄGIGE DOKUMENTE			
Kategorie	Kennzeichnung des Dokuments mit Angabe, soweit erforderlich, der maßgeblichen Teile	Betrifft Anspruch	KLASSIFIKATION DER ANMELDUNG (Int.Cl.6)
A	PATENT ABSTRACTS OF JAPAN vol. 012, no. 146 (E-605), 6.Mai 1988 & JP 62 262376 A (MITSUBISHI ELECTRIC CORP), 14.November 1987, * Zusammenfassung *	1	H01M8/02
A	CHIMICAL ABSTRACTS, vol. 114, no. 24, 17.Juni 1991 Columbus, Ohio, US; abstract no. 232033, HIYAMA ET AL: "Molten-carbonate fuel cells" XP000283481 * Zusammenfassung *	1	
A	PATENT ABSTRACTS OF JAPAN vol. 018, no. 277 (E-1554), 26.Mai 1994 & JP 06 052868 A (SUMITOMO SPECIAL METALS CO LTD), 25.Februar 1994, * Zusammenfassung *	1	RECHERCHIERTE SACHGEBIETE (Int.Cl.6)
A	DE 195 41 187 C (MOTOREN TURBINEN UNION) * Spalte 6, Zeile 52 - Zeile 67; Ansprüche 1,6 *	1,2,4,7	H01M
Der vorliegende Recherchenbericht wurde für alle Patentansprüche erstellt			
Recherchenort <b>DEN HAAG</b>		Abschlußdatum der Recherche <b>2. Januar 1998</b>	Prüfer <b>D'hondt, J</b>
KATEGORIE DER GENANNTEN DOKUMENTE X : von besonderer Bedeutung allein betrachtet Y : von besonderer Bedeutung in Verbindung mit einer anderen Veröffentlichung derselben Kategorie A : technologischer Hintergrund O : nichtschriftliche Offenbarung P : Zwischenliteratur		T : der Erfindung zugrunde liegende Theorien oder Grundsätze E : älteres Patentedokument, das jedoch erst am oder nach dem Anmeldedatum veröffentlicht worden ist D : in der Anmeldung angeführtes Dokument L : aus anderen Gründen angeführtes Dokument & : Mitglied der gleichen Patentfamilie, übereinstimmendes Dokument	

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Europäisches  
Patentamt

EUROPÄISCHER RECHERCHENBERICHT

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EINSCHLÄGIGE DOKUMENTE			
Kategorie	Kennzeichnung des Dokuments mit Angabe, soweit erforderlich, der maßgeblichen Teile	Betrifft Anspruch	KLASSIFIKATION DER ANMELDUNG (Int.Cl.6)
A	CHEMICAL ABSTRACTS, vol. 122, no. 4, 23. Januar 1995 Columbus, Ohio, US; abstract no. 36103, SATO ET AL: "Coating of stainless steel parts for resistance to corrosion of molten salt" XP002049497 * Zusammenfassung * & JP 06 260 178 A (KAWASAKI HEAVY IND) 16. September 1994 & PATENT ABSTRACTS OF JAPAN vol. 18, no. 658 (E-1643), 13. Dezember 1994 JP * Zusammenfassung * & DATABASE WPI Derwent Publications Ltd., London, GB; AN 94-336105 * Zusammenfassung *	1	
A	WO 97 23006 A (KERNFORSCHUNGSANLAGE JUELICH ;QUADAKKERS WILLEM J (NL)) ---		RECHERCHIERTE SACHGEBIETE (Int.Cl.6)
D,A	DE 44 10 711 C (KERNFORSCHUNGSANLAGE JUELICH) -----		
Der vorliegende Recherchenbericht wurde für alle Patentansprüche erstellt			
Recherchenort DEN HAAG		Abchlußdatum der Recherche 2. Januar 1998	Prüfer D'hondt, J
KATEGORIE DER GENANNTEN DOKUMENTE X : von besonderer Bedeutung allein betrachtet Y : von besonderer Bedeutung in Verbindung mit einer anderen Veröffentlichung derselben Kategorie A : technologischer Hintergrund O : nichtschriftliche Offenbarung P : Zwischenliteratur		T : der Erfindung zugrunde liegende Theorien oder Grundsätze E : älteres Patentdokument, das jedoch erst am oder nach dem Anmeldedatum veröffentlicht worden ist D : in der Anmeldung angeführtes Dokument L : aus anderen Gründen angeführtes Dokument & : Mitglied der gleichen Patentfamilie, übereinstimmendes Dokument	

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(71)Applicant : HONDA MOTOR CO LTD

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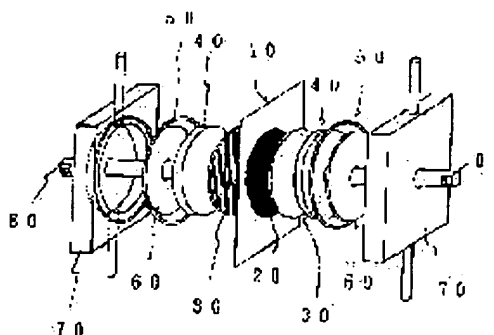
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## (54) COLLECTOR FOR FUEL CELL AND FUEL CELL USING THIS COLLECTOR

### (57)Abstract:

**PURPOSE:** To provide smallness in internal resistance and excellence in corrosion resistance and further to improve hydrogen adsorbing power by coating a surface of a collector with a metal excellent in conductivity and corrosion resistance.

**CONSTITUTION:** A fuel cell has an electrolyte film, positive electrode and a negative electrode 20 in both sides of the film, collector 30 in the outside of the electrode 20 and a collector terminal 40 brought into contact with this collector as a single cell. Here, a surface of the collector 30 consisting of porous carbon sintered material or the like is coated with a metal of platinum, gold, iridium, etc., excellent in conductivity and corrosion resistance to about 0.05 to 2 $\mu$ m by a sputtering method or the like. In this way, a sum of contact resistances between the collector 30 and the terminal 40 and between the collector and an electrode and resistance of the collector 30 itself is reduced to about 1/2. Since a metal is advanced into an interface of the collector and a catalytic layer, also hydrogen adsorbing power is improved.



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2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Industrial Application] this invention relates to the charge collector for fuel cells which can reduce internal resistance, and the fuel cell using this charge collector.

[0002]

[Description of the Prior Art] The topia block diagram of a fuel cell is shown in drawing 1 . The unit cell which consists of an electrode 20 of the anode prepared in the both sides by the electrolyte film and a cathode is inserted into a charge collector 30, a sign 10 is connected to each charge collector current collection terminal 40, one cell is constituted, and the laminating of this cell is carried out through separator. In addition, the electrode holder of the product [ sign / the flat-package-type packing and the sign 70 / which the O ring with which a sign 50 consists of a fluororubber (for example, Du Pont Viton), and a sign 60 turn into from a fluororubber (for example, Du Pont Viton) ] made from stainless steel (for example, SUS304), and a sign 80 are terminals. In order to make it the oxidation-reduction reaction of fuel gas (for example, hydrogen) and oxidizer gas (for example, oxygen) occur on each charge collector, the slot for passage on the gas is formed in the charge collector in many cases.

[0003] Reactant gas consists of fuel gas and oxidizer gas, is supplied from the passage of a charge collector, and as a result of such supply of reactant gas, an electron occurs with advance of an electrochemical reaction and it generates electrical energy by taking out this electron from an external circuit.

[0004] At this time, contact resistance is between a charge collector, an electrode, and a charge collector and a current collection terminal, and it becomes the cause by which this enlarges internal resistance of the whole fuel cell. Moreover, since it is exposed to fuel gas and oxidizer gas, although this charge collector must be the thing excellent in corrosion resistance, in the conventional thing, this point is insufficient. On the other hand, although the improvement in the hydrogen adsorption capacity in an electrode and a charge collector interface leads to the improvement in a performance of a cell, an attempt which has an effect in this is not made conventionally.

[0005]

[Problem(s) to be Solved by the Invention] this invention offers the charge collector whose hydrogen adsorption capacity was made against the background of the above Prior arts, could make internal resistance small, and was excellent in corrosion resistance, and improved, and the fuel cell using this charge collector.

[0006]

[Means for Solving the Problem] this invention offers the charge collector for fuel cells which comes to coat the metal which is excellent in conductivity and corrosion resistance on the surface of a charge collector, and the fuel cell using this charge collector.

[0007] In this invention, although especially the quality of the material, a configuration, etc. of a charge collector are not limited, a porous-carbon sintered compact, a carbon paper, its carbon cross, etc. are desirable, for example. moreover, platinum, gold, iridium, a rhodium, a ruthenium, and palladium are mentioned, and independent in these, although what is excellent in conductivity and corrosion resistance is used as a metal with which coating is presented -- or two or more sorts are combined and it is used

[0008] As the method of coating, the sputtering method, a vacuum deposition, or plating is mentioned, and a metal membrane is stuck and formed in a current collection body surface by doing in this way and coating. The thickness of a metal layer has desirable about 0.05-2 micrometers.

[0009] Thus, since the metal excellent in conductivity and corrosion resistance is used for the charge collector which had the metal coated, it can make internal resistance small and is excellent also in corrosion resistance. Moreover, since a metal enters into the interface of a charge collector and a catalyst bed, hydrogen adsorption capacity also improves.

[0010] Next, the fuel cell of this invention is characterized by using the above charge collectors, and has a current collection terminal as one cell in contact with a charge collector and this on the outside of a positive electrode and a negative electrode at an electrolyte film and its both sides in a positive electrode, a negative electrode, and a pan. What transposed the charge collector of the fuel cell shown, for example in drawing 1 to the charge collector of this invention as a fuel cell of this invention can be mentioned. And the laminating of this cell is usually carried out through separator.

[0011] In addition, in this invention, as an electrolyte film, if a solid-state polyelectrolyte film is used, it is effective. As this solid-state polyelectrolyte film, a polyperfluoro sulfo nick acid etc. is desirable. Moreover, the thickness of an electrolyte film is about 50-200 micrometers about. It may be the feature that the charge collector is coated with the metal which is excellent in

electric conduction and corrosion resistance, as long as contact resistance is reduced by this, it excels in corrosion resistance and this feature is employed efficiently, the fuel cell of this invention may not be limited to an above-mentioned example, and the fuel cell of what form is sufficient as it.

[0012] In this invention, since the metal which is excellent in conductivity and corrosion resistance at a current collection body surface is coated, the contact resistance in the contact surface between a charge collector, an electrode, and a charge collector and a charge collector terminal can be reduced, and internal resistance of a cell can be made small, and corrosion-resistant improvement can also be aimed at. Furthermore, a metal can enter into the interface of a charge collector and an electrode, and, thereby, hydrogen adsorption capacity can be raised.

[0013] Although an example is given to below and this invention is explained, this invention is not limited to these examples. The charge collector which consists of an example 1 porosity carbon board was coated with platinum so that it might become the thickness of 0.1 micrometers by sputtering. It generated electricity by constructing a cell as shown in drawing 2 using this charge collector. In addition, in drawing 2, each sign is the same as that of drawing 1, for an electrode and a sign 30, a charge collector and the sign 40 of an electrolyte film and a sign 20 are charge collector terminals, and a sign 10 is the metal coating film with which this charge collector 30 was coated with the sign 90.

[0014] At this time, they are between charge collector-current collection terminals and a charge collector. - The sum (it is only hereafter described as "resistance") R of inter-electrode contact resistance and the own resistance of a charge collector is expressed with  $R=(V1-V2)/A$ . Then, they are R1 and resistance when not coating about the resistance when coating R2. It carries out, current density is considered and it is  $R1 / R2$ . It measured. A result is shown in drawing 3. From drawing 3, by coating a charge collector with a metal shows that resistance decreases about to 1/2.

[0015]

[Effect of the Invention] the hydrogen adsorption capacity [ in / a charge collector-catalyst bed interface / further / things can be carried out and ] which corrosion resistance improves and makes internal resistance small since the charge collector of this invention has coated the metal excellent in conductivity and corrosion resistance -- improvement -- things are made

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[Translation done.]

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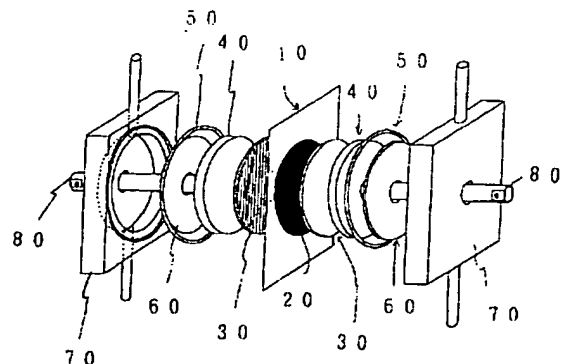
(74)代理人 弁理士 白井 重隆

(54)【発明の名称】 燃料電池用集電体およびそれを用いた燃料電池

(57)【要約】

【構成】 集電体の表面に、導電性と耐蝕性に優れる金属をコーティングした集電体。

【効果】 接触抵抗が低減し、内部抵抗の低減された電池が得られる。また、集電体の耐蝕性が向上する。さらに、集電体と触媒層界面での水素吸着能の向上が図れ、電池の性能が向上する。



## 【特許請求の範囲】

【請求項1】 集電体の表面に導電性と耐蝕性に優れる金属をコーティングしてなる燃料電池用集電体。

【請求項2】 金属が、白金、金、イリジウム、ロジウム、ルテニウムおよびパラジウムの群から選ばれた少なくとも1種である請求項1記載の集電体。

【請求項3】 請求項1記載の集電体を用いた燃料電池。

## 【発明の詳細な説明】

【0001】

【産業上の利用分野】本発明は、内部抵抗を低減できる燃料電池用集電体およびこの集電体を用いた燃料電池に関する。

【0002】

【従来の技術】燃料電池の斜視構成図を図1に示す。符号10は、電解質膜でその両側に設けられたアノードおよびカソードの電極20からなる単位電池が集電体30に挟まれ、それぞれの集電体に集電端子40接続され、1つのセルが構成され、このセルがセパレータを介して積層されている。なお、符号50はフッ素ゴム（例えば、デュボン社製バイトン）からなるOリング、符号60はフッ素ゴム（例えば、デュボン社製バイトン）からなる平型バックリング、符号70はステンレス（例えば、SUS304）製のホルダー、符号80は端子である。各集電体上で燃料ガス（例えば、水素）と酸化剤ガス（例えば、酸素）との酸化還元反応が起こるようにするため、集電体にはガスの流路用溝が形成されていることが多い。

【0003】反応ガスは、燃料ガスと酸化剤ガスからなり、集電体の流路から供給され、このような反応ガスの供給の結果、電気化学的反応の進行にともない電子が発生し、この電子を外部回路から取り出すことにより、電気エネルギーを発生する。

【0004】このときに、集電体と電極および集電体と集電端子間に、接触抵抗があり、これが燃料電池全体の内部抵抗を大きくしてしまう原因となる。また、この集電体は、燃料ガス、酸化剤ガスにさらされるので、耐蝕性に優れたものでなければならないが、従来のものではこの点で不充分である。一方、電極と集電体界面での水素吸着能の向上は、電池の性能向上につながるが、従来、これに効果のあるような試みはなされていない。

【0005】

【発明が解決しようとする課題】本発明は、以上のような従来の技術を背景になされたものであり、内部抵抗を小さくでき、耐蝕性に優れ、かつ水素吸着能が向上した集電体およびこの集電体を用いた燃料電池を提供するものである。

【0006】

【課題を解決するための手段】本発明は、集電体の表面に導電性と耐蝕性に優れる金属をコーティングしてなる

燃料電池用集電体、およびこの集電体を用いた燃料電池を提供するものである。

【0007】本発明において、集電体の材質および形状などは、特に限定されるものではないが、例えば多孔質炭素炭結体、カーボンペーパー、カーボクロスなどが好ましい。また、コーティングに供される金属としては、導電性と耐蝕性に優れるものが用いられるが、白金、金、イリジウム、ロジウム、ルテニウム、パラジウムが挙げられ、これらを単独あるいは2種以上組み合わせ用いられる。

【0008】コーティングの方法としては、スパッタリング法、蒸着法、あるいはメッキ法などが挙げられ、このようにしてコーティングすることにより、金属膜が集電体表面に密着して形成される。金属層の厚みは、0.05～2μm程度が好ましい。

【0009】このようにして金属をコーティングされた集電体は、導電性、耐蝕性に優れた金属を用いているので、内部抵抗を小さくすることができ、耐蝕性にも優れる。また、集電体と触媒層の界面に金属が入り込むため、水素吸着能も向上する。

【0010】次に、本発明の燃料電池は、上記のような集電体を用いたことを特徴とし、電解質膜、その両側に正極と負極、さらに正極、負極の外側に集電体およびこれと接して集電端子を1つのセルとして有する。本発明の燃料電池としては、例えば図1に示す燃料電池の集電体を本発明の集電体に置き換えたものを挙げることができる。そして、通常、このセルは、セパレータを介して積層される。

【0011】なお、本発明において、電解質膜としては、固体高分子電解質膜を用いると効果的である。この固体高分子電解質膜としては、ポリパーフルオロスルフォニック酸などが好ましい。また、電解質膜の膜厚は、おおよそ50～200μm程度である。本発明の燃料電池は、集電体が導電および耐蝕性に優れる金属でコーティングされていることが特徴であり、これにより接触抵抗が低減され、耐蝕性に優れているのであり、この特徴が生かされているものであれば、上述の例に限定されるものではなく、どのような形式の燃料電池でもよい。

【0012】本発明においては、集電体表面に導電および耐蝕性に優れる金属をコーティングしているため、集電体と電極および集電体と集電端子間の接触面で接触抵抗を低減することができ、電池の内部抵抗を小さくすることができ、また耐蝕性の向上も図ることができる。さらに、集電体と電極の界面に金属が入り込み、これにより水素吸着能を向上させることができる。

【0013】以下に実施例を挙げ、本発明を説明するが、本発明はこれらの実施例に限定されるものではない。

## 実施例1

多孔質カーボン板からなる集電体に、白金をスパッタリ

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ングにより厚み0.1  $\mu\text{m}$ になるようにコーティングした。この集電体を用い、図2に示すようなセルを組み、発電を行った。なお、図2において、各符号は図1と同様であり、符号10は電解質膜、符号20は電極、符号30は集電体、符号40は集電体端子であり、また符号90はこの集電体30にコーティングされた金属コーティング膜である。

【0014】このとき、集電体-集電端子間、および集電体-電極間の接触抵抗と集電体自身の抵抗の和（以下、単に「抵抗」と記す） $R$ は、 $R = (V_1 - V_2) / A$ で表される。そこで、コーティングしたときの抵抗を $R_1$ 、コーティングしないときの抵抗を $R_2$ とし、電流密度を考えて $R_1 / R_2$ を測定した。結果を図3に示す。図3より、集電体に金属をコーティングすることにより、抵抗が1/2程度に減少することが分かる。

【0015】

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【発明の効果】本発明の集電体は、導電性と耐蝕性に優れた金属をコーティングしているため、耐蝕性が向上し、内部抵抗を小さくことができ、さらには集電体-触媒層界面における水素吸着能を向上することができる。

【図面の簡単な説明】

【図1】燃料電池の斜視構成図である。

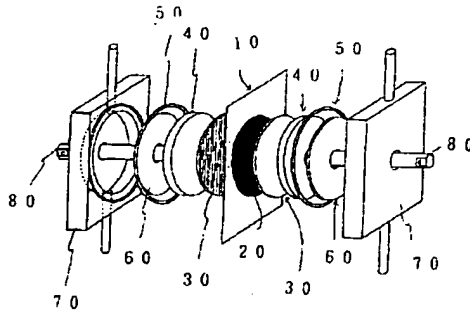
【図2】実施例1で用いられるセルの断面図である。

【図3】実施例1における測定結果を示すグラフである。

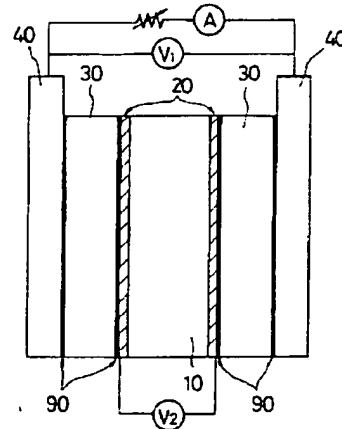
【符号の説明】

- 10 電解質膜
- 20 電極
- 30 集電体
- 40 集電端子
- 90 金属コーティング膜

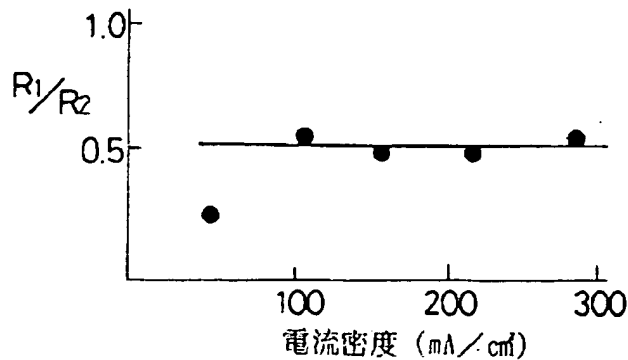
【図1】



【図2】



【図3】



US-PAT-NO: 6454978

DOCUMENT-IDENTIFIER: US 6454978 B1

TITLE: Process for making fuel cell plates

----- KWIC -----

The '370 patent discloses a new fluid flow field plate construction consisting of a stencil layer and a separator layer. The separator and stencil layers are formed of flexible graphite foil sheets having a thickness between about 0.003 inches and about 0.030 inches. Another patent, U.S. Pat. No. 5,521,018 ('018), discloses the concept of embossing a fluid flow field plate such as electrically conductive graphite foil sheet material. Other materials being sufficiently soft so as to permit embossing include porous electrically conductive sheet materials, such as carbon fibre paper, corrosive resistant metals, such as niobium; somewhat corrosive resistant material, such as magnesium or copper particularly when plated with noble metals such as gold or platinum to render them unreactive; and composite materials composed of corrosive metal powder, a base metal powder plated with corrosive resistant metal, and/or other chemically inert electrically conductive powders such as graphite and boron carbide bonded together with a suitable binder to produce a compressible electrically conductive sheet material. The embossing step is accomplished using a die where the channels are generally U-shaped or V-shaped in cross section. The '018 patent discloses that "the graphite foil sheet is embossed at an embossing pressure sufficient to impart into the compressible



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(19) **United States**(12) **Patent Application Publication** (10) Pub. No.: **US 2001/0006702 A1**  
Hisada et al. (43) Pub. Date: **Jul. 5, 2001**(54) **METHOD FOR DEPOSITING METAL  
HAVING HIGH CORROSION RESISTANCE  
AND LOW CONTACT RESISTANCE  
AGAINST CARBON ON SEPARATOR FOR  
FUEL CELL****Publication Classification**(51) Int. Cl.<sup>7</sup> ..... B05D 1/12; B05D 5/00  
(52) U.S. Cl. .... 427/115; 427/126.5; 427/191;  
427/427(75) Inventors: **Wataru Hisada, Nagoya-shi (JP);  
Hiroshi Kihira, Futsu City (JP)**(57) **ABSTRACT**Correspondence Address:  
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A method for depositing a metal having a high corrosion resistance and a low contact resistance against carbon to a separator for a fuel cell enabling provision of an inexpensive separator for a fuel cell by depositing a metal having a high corrosion resistance and a low contact resistance against carbon to the surface of a metal conveniently by simple equipment while using as a preform a metal such as stainless steel or aluminum as a material having a high productivity and low price and in addition capable of reducing the weight by making the sheet thickness thin, comprising projecting to a separator of a unit cell for forming the fuel cell a solid plating material comprised of core particles having a higher hardness than the separator and coated with a metal having a high corrosion resistance and a low contact resistance against carbon so as to compulsorily deposit the metal coated on this solid plating material to the separator.

(73) Assignee: (1) **Sintobrator, Ltd.**, (2) **Nippon Steel Corporation.**(21) Appl. No.: **09/748,490**(22) Filed: **Dec. 26, 2000**(30) **Foreign Application Priority Data**Dec. 27, 1999 (JP) ..... 11-369817  
Jun. 29, 2000 (JP) ..... 2000-196526

Fig. 1

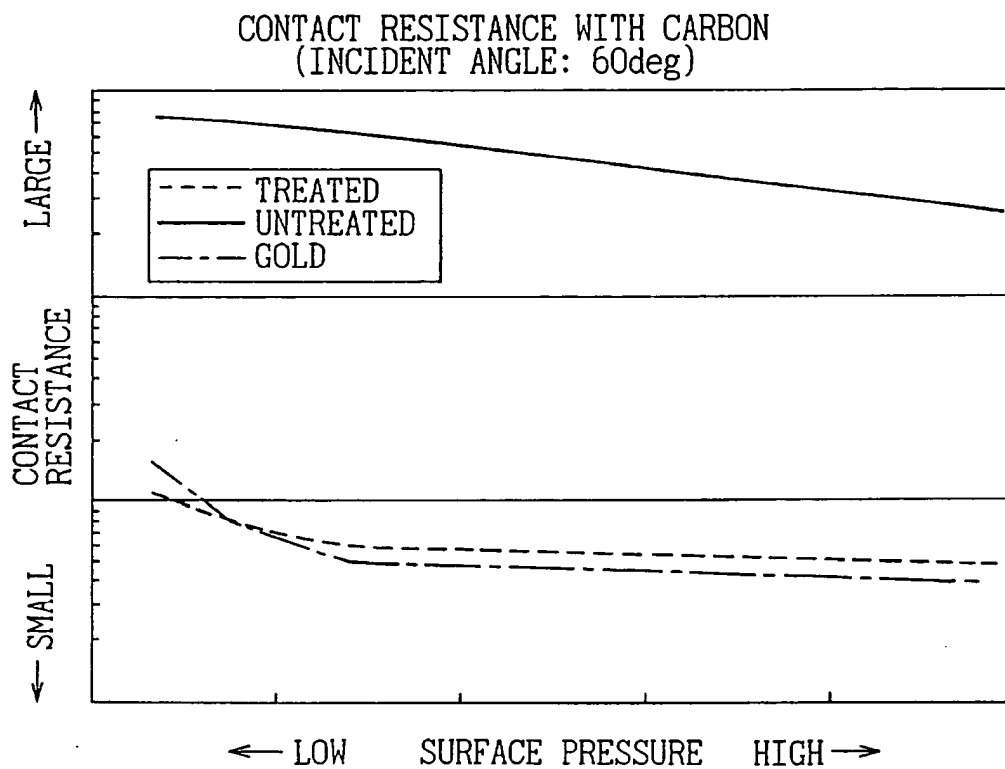
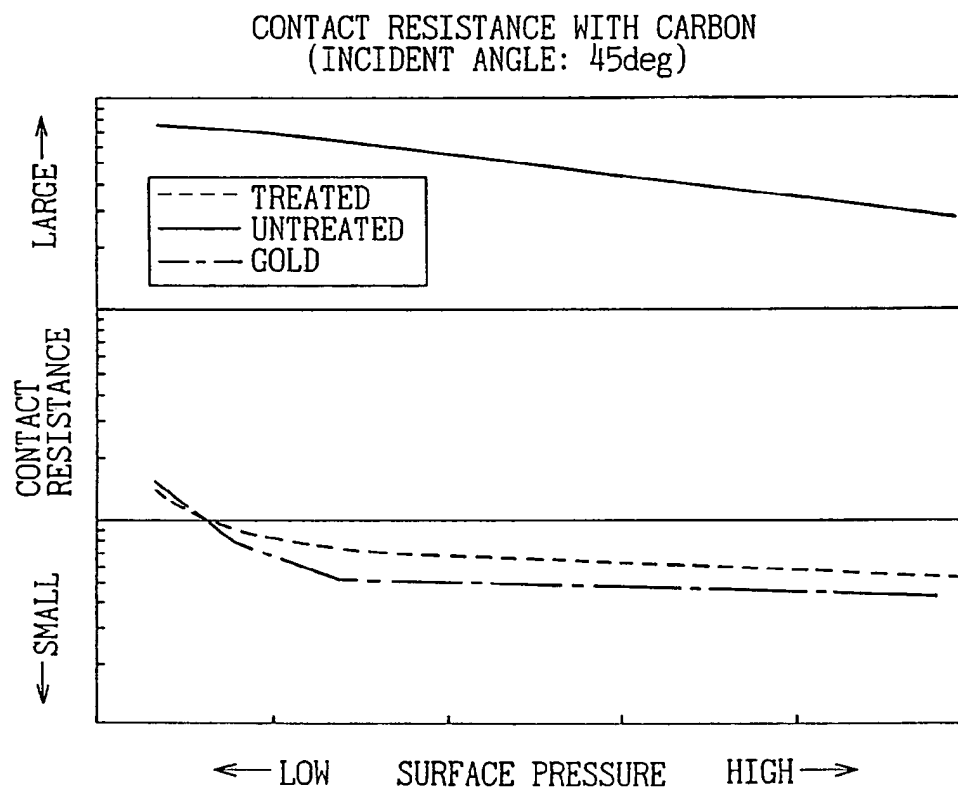




Fig. 2



# METHOD FOR DEPOSITING METAL HAVING HIGH CORROSION RESISTANCE AND LOW CONTACT RESISTANCE AGAINST CARBON ON SEPARATOR FOR FUEL CELL

## BACKGROUND OF THE INVENTION

### [0001] 1. Field of the Invention

[0002] The present invention relates to a method for depositing a metal having a high corrosion resistance and a low contact resistance against carbon to a separator of a unit cell for forming a fuel cell.

### [0003] 2. Description of the Related Art

[0004] As a fuel cell, there are a solid polymer type fuel cell, a phosphoric acid type fuel cell, a molten carbonate type fuel cell, an alkali type fuel cell, etc. These fuel cells use different electrolytes, but acquire electromotive force by an electrochemical reaction of hydrogen and oxygen. A fuel cell is formed by stacked unit batteries (cells) and a separator for electrically connecting cells and for separating a reaction gas fed to the cells disposed between adjoining cells.

[0005] As this separator, a carbon material has been mainly used. With carbon, however, the required separator shape has been obtained by a shaving from a preform or pressurized powder shaping or the like. Accordingly, the productivity was low and the manufacturing cost high. Further, a separator made of titanium has been partially used as a separator made of metal, but titanium is an expensive material, so there is a limit in lowering of costs.

[0006] Therefore, a separator using as a preform a metal such as stainless steel or aluminum, which are materials having a high productivity and a low cost and which enable of reduction a weight by making the sheet thickness smaller has been proposed in Japanese Unexamined Patent Publication (Kokai) No. 10-228914 etc. When using a metal as a preform, in order to lower the contact resistance against the carbon electrode or carbon paper forming a current collector and then impart corrosion resistance, it is necessary to plate it with a precious metal. This plating must be done immediately after removal of a passive film unlike with usual plating. Therefore, in a case of a wet type, there was a problem in that many steps such as degreasing, washing, surface activation, washing, plating, washing, and drying had to be passed. The hardware per se became large in size in physical vapor deposition (PVD) and chemical vapor deposition (CVD).

## SUMMARY OF THE INVENTION

[0007] An object of the present invention is to provide a method for depositing a metal having a high corrosion resistance and a low contact resistance against carbon to a separator for a fuel cell enabling provision of an inexpensive separator for a fuel cell by depositing a metal having a high corrosion resistance and a low contact resistance against carbon to the surface of a metal conveniently by simple equipment while using as a preform a metal such as stainless steel or aluminum as a material having a high productivity and low price and in addition capable of reducing the weight by making the sheet thickness thin.

[0008] According to the present invention, there is provided a method of depositing a metal having a high corro-

sion resistance and a low contact resistance against carbon on a separator for a fuel cell, the method comprising the steps of projecting to a separator of a unit cell for forming the fuel cell a solid plating material comprised of core particles having a higher hardness than the separator and coated with a metal having a high corrosion resistance and a low contact resistance against carbon so as to compulsorily deposit the metal coated on this solid plating material to the separator.

[0009] Preferably, a projection velocity of the solid plating material to the separator is made 20 to 100 m/sec.

[0010] Preferably, the projection of the solid plating material to the separator is performed by a flow of dry air. Alternatively, the projection of the solid plating material is performed by a rotating impeller. Alternatively, the projection of the solid plating material is performed by a flow of water. Alternatively, the projection of the solid plating material is performed by a flow of inert gas.

[0011] More preferably, the core particles of the solid plating material have a particle size of 30 to 300  $\mu\text{m}$ , a true specific gravity of 2 to 15, and a hardness of 400/2000 Hv.

[0012] Still more preferably, the core particles of the solid plating material are made of hard metal.

[0013] Still more preferably, the metal having a high corrosion resistance and a low contact resistance against carbon to be coated on the core particles of the solid plating material is a single metal or an alloy.

[0014] Still more preferably, the metal having a high corrosion resistance and a low contact resistance against carbon to be coated on the core particles of the solid plating material is at least one of gold, silver, copper, and nickel.

[0015] Still more preferably, the carbon contact resistance value is not more than 20  $\text{m}\Omega\cdot\text{cm}^2$  at a contact pressure of at least 1  $\text{kg.f/cm}^2$ .

## BRIEF DESCRIPTION OF THE DRAWINGS

[0016] These and other objects and features of the present invention will become clearer from the following description of the preferred embodiments given with reference to the accompanying drawings, wherein:

[0017] FIG. 1 is a graph of the relationship between face pressure and a contact resistance value in experimental results of Example 1 of the present invention and a comparative specimen and

[0018] FIG. 2 is a graph of the relationship between face pressure and a contact resistance value in experimental results of Example 2 of the present invention and a comparative specimen.

## DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0019] The present invention provides a method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon on a separator for a fuel cell comprising the steps of projecting to a separator of a unit cell for forming the fuel cell a solid plating material comprised of core particles having a higher hardness than the separator coated with a metal having a high corrosion

resistance and a low contact resistance against carbon so as to compulsorily deposit the metal coated on this solid plating material to the separator.

[0020] According to the method of the present invention, the solid plating material has a higher hardness than the separator material. Therefore, if the solid plating material strikes upon the separator material, first the passivation film at the surface of the separator material is destroyed and the true surface of the preform appears. At the same time, adequate surface roughening (anchor pattern effect) occurs, plastic deformation mechanically occurs in the coating material of the solid plating material by this anchor portion (scratching effect), the coating material is compulsorily deposited to the preform true surface of the separator, the deposited coating material is rolled by the projection pressure by the core particles, and thus a uniform coating layer is formed on the surface of the separator material.

[0021] Note that if instead of using the solid plating material as described above, a solid plating material with all particles made of a soft material such as gold or silver is projected, even if it strikes the separator material, it cannot sufficiently destroy the passivation film at the surface of the separator material. In addition, the anchor pattern effect cannot be obtained. Further, a precious metal such as gold and the silver is expensive. Therefore, the destruction of the passivation film and the formation of the low contact resistance layer aimed at by the present invention cannot be simultaneously conveniently carried out.

[0022] Further, in the present invention, the projection velocity of the solid plating material is selected within a range of 20 to 100 m/sec according to the material of the core particles. The reason for this is that there is an optimum combination of the thickness and strength of the passivation film, the thickness of the preform and the hardness and the true specific gravity of the core particles according to the material of the separator. For example, in the case where the core particles are hard metal, they have a high hardness and a high specific gravity, so can sufficiently function even at a low velocity, but preferably it is about 20 to 40 m/sec when considering the running cost and deformation of the separator or the like. In contrast, when the core particles have a medium hardness and a low specific gravity such as glass beads, efficient processing cannot be carried out unless the velocity is set higher than the former within a range up to 100 m/sec.

[0023] Further, in the present invention, the acceleration medium of the solid plating material may be any of a flow of dry air, an impeller, a flow of water, and a flow of inert gas so far as the flow rate and the projection velocity of the solid plating material can be controlled, but a flow of dry air is most suitable when considering the flow rate of the solid plating material, the control of the projection velocity, economy, and workability.

[0024] Note that if the solid plating material is accelerated and strikes the separator surface as in the present invention, there is also a cleaning effect, so the method can be executed by omitting a pretreatment step in the case of a slightly contaminated separator surface.

[0025] Below, the present invention will be explained further using examples given in comparison with comparative specimens.

#### [0026] Known Specimens

[0027] First, carbon paper was held between two specimens of gold plates having the same area as the carbon paper. Various loads were applied between the two gold plates to change the face pressure, a constant current was supplied between the two gold plates, and the voltage at that time was measured. Then, the current density was calculated from the area of each specimen and the face resistivity at each face pressure was calculated from the measurement result of each specimen so as to obtain the contact resistance value between the gold and the carbon paper.

[0028] Next, carbon paper of the same area as the specimen was held between a specimen of stainless steel plate (JIS SUS316) having the same area as the gold plates described above and polished to a mirror finish and a gold plate the same as one of the gold plates described above. A contact current was supplied between this stainless steel plate and the gold plate and the voltage at that time was measured. The contact resistance value between the stainless steel plate and the carbon paper was calculated from the results thereof in a similar way to that described above.

#### EXAMPLE 1

[0029] A solid plating material obtained using hard metal particles of 100  $\mu$ m size as core particles, using gold as the coating material, and directly plating this gold on the surface of the core particles was obtained for use in the present invention. On the other hand, as the preform acting as the separator, use was made of a test piece for measuring the contact resistance obtained by mirror polishing a disk made of stainless steel plate (JIS SUS316) having a diameter of 30 mm and a thickness of 4 mm. The solid plating material was accelerated toward the surface with an air pressure of 0.4 MPa, projected at conditions of a velocity of 35 m/min and an incident angle of 60 degrees, and struck the surface of the test piece for measuring the contact resistance to obtain a test piece with a uniform coating of gold formed on the surface of the test piece.

[0030] When measuring the contact resistance value of this test piece and the carbon paper at a contact pressure of 1 kg.f/cm<sup>2</sup> and calculating by a method similar to that of the case of when preparing the known specimens, the result became less than 20 m $\Omega$ .cm<sup>2</sup> or about 1/100 of the contact resistance value between the untreated stainless steel plate of the known specimen and carbon paper at a contact pressure of 1 kg.f/cm<sup>2</sup>.

#### EXAMPLE 2

[0031] Use was made of a solid plating material and a test piece for measuring the contact pressure serving as the preform acting as the separator the same as those of Example 1. The solid plating material was accelerated toward the surface of this test piece with an air pressure of 0.4 MPa and struck at a velocity of 35 m/min and an incident angle of 45 degrees. As a result, a test piece for measuring the contact resistance with a uniform coating of gold formed on the surface of the test piece was obtained.

[0032] When measuring the contact resistance value of this test piece and the carbon paper at a contact pressure of 1 kg.f/cm<sup>2</sup> and calculating by a method similar to that of the

case of when preparing the known specimens, the result became less than  $20 \text{ m}\Omega\cdot\text{cm}^2$  or about  $1/100$  of the contact resistance value between the untreated stainless steel plate used as the known specimen and carbon paper at a contact pressure of  $1 \text{ kg.f/cm}^2$ .

[0033] The contact resistance values between the gold described above as the known specimens and the carbon at the different face pressures and the contact resistance values between the untreated stainless steel and carbon at the different face pressures and also the contact resistance values at the different face pressures in Example 1 and Example 2 treated by the method of the present invention are shown in FIG. 1 and FIG. 2.

[0034] According to experiments, the contact resistance value obtained by the processing by the method of the present invention was  $3$  to  $5 \Omega\cdot\text{cm}^2$  at the maximum face pressure  $10 \text{ kg.f/cm}^2$  of the present test. This is about  $1/100$  with respect to the contact resistance value between the untreated stainless steel explained as the known specimen and carbon at the same contact pressure, i.e.,  $300$  to  $500 \text{ m}\Omega\cdot\text{cm}^2$  and substantially equivalent to the contact resistance value of  $3$  to  $4 \text{ m}\Omega\cdot\text{cm}^2$  between the gold explained as the known specimens and carbon at the same contact pressure.

[0035] As apparent from the above explanation, the present invention has the advantages that when forming gold or another metal coating of a high corrosion resistance and a low contact resistance against carbon on a preform made of a metal having a high productivity and low cost such as stainless steel or aluminum used as the separator of a unit cell for forming a fuel cell, there is the advantage that the processing becomes possible at a high productivity and with a simple method without wet type plating which requires a complex process and large system and in addition requires trouble for processing the waste liquor and wastewater.

[0036] Accordingly, the present invention greatly contributes to development of the industry as a method for depositing a metal having a high corrosion resistance and a low contact resistance against carbon on the surface of stainless steel, aluminum, or another metal by a simple system and thereby enables provision of a cheap separator for a fuel cell.

[0037] While the invention has been described by reference to specific embodiments chosen for purposes of illustration, it should be apparent that numerous modifications could be made thereto by those skilled in the art without departing from the basic concept and scope of the invention.

What is claimed is

1. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon on a separator for a fuel cell, the method comprising the steps of projecting to a separator of a unit cell for forming the fuel cell a solid plating material comprised of core particles

having a higher hardness than the separator and coated with a metal having a high corrosion resistance and a low contact resistance against carbon so as to compulsorily deposit the metal coated on this solid plating material to the separator.

2. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in claim 1, wherein a projection velocity of the solid plating material to the separator is  $20$  to  $100 \text{ m/sec}$ .

3. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in claim 1 or 2, wherein the projection of the solid plating material to the separator is performed by a flow of dry air.

4. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in claim 1 or 2, wherein the projection of the solid plating material is performed by a rotating impeller.

5. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in claim 1 or 2, wherein the projection of the solid plating material is performed by a flow of water.

6. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in claim 1 or 2, wherein the projection of the solid plating material is performed by a flow of inert gas.

7. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in any one of claims 1 to 6, wherein the core particles of the solid plating material have a particle size of  $30$  to  $300 \mu\text{m}$ , a true specific gravity of  $2$  to  $15$ , and a hardness of  $400/2000 \text{ Hv}$ .

8. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in any one of claims 1 to 7, wherein the core particles of the solid plating material are made of hard metal.

9. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in any one of claims 1 to 8, wherein the metal having a high corrosion resistance and a low contact resistance against carbon to be coated on the core particles of the solid plating material is a single metal or an alloy.

10. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in any one of claims 1 to 9, wherein the metal having a high corrosion resistance and a low contact resistance against carbon to be coated on the core particles of the solid plating material is at least one of gold, silver, copper, and nickel.

11. A method of depositing a metal having a high corrosion resistance and a low contact resistance against carbon as set forth in any one of claims 1 to 10, wherein the carbon contact resistance value is not more than  $20 \text{ m}\Omega\cdot\text{cm}^2$  at a contact pressure of at least  $1 \text{ kg.f/cm}^2$ .

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